

(11) Publication number : **0 457 554 A2**

(12)

EUROPEAN PATENT APPLICATION

(21) Application number : **91304330.3**

(51) Int. Cl.⁵ : **C08L 23/22, F16F 7/00**

(22) Date of filing : **15.05.91**

(30) Priority : **15.05.90 JP 124666/90**

(43) Date of publication of application :
21.11.91 Bulletin 91/47

(84) Designated Contracting States :
DE FR GB IT

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(54) **Rubber composition having a large damping capacity.**

(57) Disclosed is rubber composition which can preferably be used for components requiring high damping capacity such as rubber dampers for use in structures, public works, vehicles, oversize powered machines, and heavy weight structures; and general purpose dampers for use in sound products, a variety of meters, the components of the vehicles, and the like in the case of a little amount of use. The rubber composition has a complex shear elastic modulus of 1-20 kg/cm² and a Tanδ of 0.40-1.5 at 30 °C under a frequency of 0.5 Hz ± 50 % comprising :

a) x parts by weight of a filler for use in rubber ;

b) y parts by weight of carbon black having an iodine adsorption of 20-120 mg/g ; and

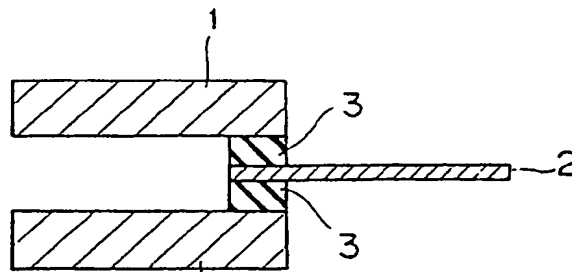
c) 100 parts by weight of polyisobutylene ;

wherein the value x and y satisfy the relation that :

$x \leq 450$, $y \leq 150$, and

$x + y \geq 100$.

Fig. 1



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The present invention relates to a rubber composition having a large damping capacity.

Recently, laminated vibrationproofing structures have been widely used as vibration isolators for constructive structures (e.g. buildings). The laminated vibrationproofing structures are introduced between the structures and the foundation to decrease transmission of earthquake vibration energy to the structures. A wide variety of shapes for these laminated vibrationproofing structures have been proposed. Rubber compositions having a large damping capacity have been used as components of these laminated vibration proofing structures.

In order to enhance damping capacity, the rubber composition contains carbon black in a large amount. Processing characteristics and excellent elongation are also imparted by addition of a large amount of softening agent to the rubber composition. Although these conventional rubber compositions have good damping capacity, a rubber composition having a constant damping capacity in a wide range of temperature (e.g., -10 to +30 °C) and excellent aging resistance is desired.

An object of the present invention is to provide a rubber composition which can preferably be used for components requiring a high damping capacity such as rubber dampers for use in structures, public works, vehicles, oversize powered machines, and heavy weight structures; and general purpose dampers for use in sound products, a variety of meters, components of vehicles, and the like in cases of a little amount of use. The rubber composition of the present invention, accordingly, has a large damping capacity, a decreased variation of elastic modulus and damping capacity at a wide temperature range (e.g., It does not crystallize at low temperature-), and an excellent aging resistance. Thus, the present invention provides a rubber composition having a complex shear elastic modulus (Hereinafter, the complex shear elastic modulus is referred to as G^* .) of 1-20 kg/cm² and a $\tan\delta$ of 0.40-1.5 at 30 °C under a frequency of 0.5 Hz \pm 50 % comprising:

- a) x parts by weight of a filler for use in rubber;
- b) y parts by weight of carbon black having an iodine adsorption of 20-120 mg/g; and
- c) 100 parts by weight of polyisobutylene;

wherein the value x and y satisfy the relation that:

$$x \leq 450, y \leq 150, \text{ and}$$

$$x + y \geq 100.$$

Fig. 1 is a sectional view of a sample which is prepared for measurement of G^* and $\tan\delta$ of the rubber composition of the present invention.

The rubber component useful in the present invention is polyisobutylene. Other rubber components such as butyl rubber, halogenated butyl rubber, natural rubber, polyisoprene, styrene-butadiene rubber, acrylonitrile-butadiene rubber, chlorosulfonated polyethylene rubber, ethylene propylene rubber, chloroprene rubber, butadiene rubber, and polynorbornene rubber can also be blended to the rubber composition of the present invention. Suitable polyisobutylene employed in the present invention has a molecular weight of not less than 300,000, preferably not less than 1,000,000 as obtained from Flory equation for melt viscosity. In the present invention, the wording "molecular weight" means "the Flory viscosity average molecular weight". If the molecular weight is less than 300,000, the damping capacity of the resulting rubber composition may be lessened due to the difficulty of mixing the large amount of carbon black and fillers in the composition.

The carbon black employed in the present invention has an iodine adsorption of 20-120 mg/g as measured by a method described in JIS K6221. If the iodine adsorption is less than 20 mg/g, the damping capacity of the resulting rubber composition may be lessened. If the iodine adsorption is greater than 120 mg/g, dispersion during the kneading process, and processing characteristics in the successive processes (e.g. extrusion, calendaring) may be lessened.

The fillers employed in the present invention may be conventional fillers for use in rubber composition in the art. Examples of suitable fillers include, but are not limited to, finely powdered talc, hard clay, surface treated calcium carbonate, calcium silicate, silica, and chinese white.

The amount of carbon black incorporated in the rubber composition of the present invention is not more than 150 parts by weight based on the 100 parts by weight of polyisobutylene. The amount of the fillers incorporated in the rubber composition of the present invention is not more than 450 parts by weight based on the 100 parts by weight of polyisobutylene. Furthermore, the combined amount of the carbon black and the fillers must be not less than 100 parts by weight. If the combined amount of the carbon black and the filler is less than 100 parts by weight, the damping capacity of the resulting rubber composition may be lessened. If greater than 150 parts by weight of the carbon black or greater than 450 parts by weight of the filler is blended, the processing characteristics of the resulting rubber composition may be lessened.

Other conventional additives such as softening agent, tackifier, oligomer, anti-aging agent, and processing aid may optionally be incorporated in the rubber composition of the present invention. Preferably, these additives are incorporated in the rubber composition of the present invention in an amount of not more than 120 parts by weight based on 100 parts by weight of the rubber components.

Since polyisobutylene has little double bonds in its main chain, it may not be vulcanized. However, in the rubber composition of the present invention, other rubber components having double bonds in their main chain may be blended. Then, the rubber composition of the present invention containing rubber components other than polyisobutylene may be vulcanized by addition of vulcanizing agents or vulcanizing accelerators.

The rubber composition of the present invention must have a G^* of 1-20 kg/cm² and a $\tan\delta$ of 0.40-1.5 at 30 °C under a frequency of 0.5 Hz \pm 50 %. $\tan\delta$ is a physical property which represents damping capacity of the material. Preferably, G^* of the rubber-like viscoelastic material employed in the present invention is 1-20 kg/cm², more preferably 3-12 kg/cm². The material having G^* less than 1 kg/cm² is inferior in damping capacity. Since the spring constant in the shear direction of the laminated rubber structures is defined by G^* and the height in vertical direction of the laminated rubber structures, if G^* is greater than 20 kg/cm², the height of the laminated rubber structures becomes large to produce buckling without difficulty. Thus, a material having G^* of not more than 20 kg/cm² is preferred.

The procedure for measurement of G^* and $\tan\delta$ of the present invention is as follows. First, in the rubber composition 3 which is held between metal plates 1, 1, another metal plate 2 is introduced as shown in Fig. 1. The metal plates 1, 1 are then fixed, the metal plate 2 is moved horizontal direction at a frequency of 0.5 Hz to obtain the value of G^* and $\tan\delta$.

The shear deformation is defined as the following formula:

$$\text{Shear deformation (\%)} = l_1/t_0 \times 100.$$

l_1 : Horizontal distance in which the metal plate 2 was moved.

t_0 : Thickness of the rubber composition 3.

EXAMPLES

The following non-limiting examples further illustrate the present invention.

Examples 1-4 and Comparative Examples 1-5

Rubber compositions were prepared according to the formulation included in the following Table 1.

The physical properties of the resulting rubber compositions are reported in Table 2. As the physical properties, G^* and $\tan\delta$ both at 30 °C and at -10 °C, G^* at -10 °C where the rubber sample had been aged for 16 hours at -10 °C, as well as a rate of change of the Mooney viscosity before and after an aging for 168 hours at 100 °C were measured.

Example 1 and Comparative Examples 1 and 2 show that the polyisobutylene rubber composition of the present invention has a decreased change of G^* in the range of temperature between 30 °C and -10 °C. This indicates that the rubber composition of the present invention does not crystallize under the abovenoted conditions. Apparent from Comparative Examples 3 and 5, damping capacity is reduced when the amounts of the carbon black and the fillers are decreased. Comparative Example 4 shows that the rubber composition containing too much carbon black and the fillers has reduced processing characteristics such as kneading, carbon dispersion, and calendering. Furthermore, the rubber composition of the present invention exhibits a decreased change of the Mooney viscosity before and after aging for 168 hours at 100 °C. This indicates that the rubber composition of the present invention has an excellent aging resistance.

Table 1

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Components

| | <u>E^{a)}₁</u> | <u>C^{b)}₁</u> | <u>C₂</u> | <u>E₂</u> | <u>E₃</u> | <u>E₄</u> | <u>C₃</u> | <u>C₄</u> | <u>C₅</u> |
|-----------------------|-----------------------------------|-----------------------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|
| 10 PIB ¹⁾ | 100 | - | - | 100 | 100 | 100 | 100 | 100 | 100 |
| NR ²⁾ | - | 100 | - | - | - | - | - | - | - |
| SBR ³⁾ | - | - | 100 | - | - | - | - | - | - |
| 15 CB26 ⁴⁾ | 70 | 70 | 70 | - | - | - | - | 180 | - |
| CB110 ⁵⁾ | - | - | - | 70 | 50 | 120 | 30 | - | 60 |
| 20 PT ⁶⁾ | 85 | 85 | 85 | 85 | 405 | 215 | 40 | 200 | 20 |
| SA ⁷⁾ | 22 | 22 | 22 | 29 | 58 | 56 | 18 | 65 | 20 |
| StA ⁸⁾ | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 |
| 25 TiC ⁹⁾ | 1.2 | 1.2 | 1.2 | 1.2 | 6 | 3 | 0.7 | 3 | - |

a) Example

b) Comparative Example

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1) Polyisobutylene

2) Natural rubber

3) Styrene-butadiene rubber

4) Carbon black having an iodine adsorption of 26 mg/g

5) Carbon black (LS-ISAF) having an iodine adsorption of 110 mg/g

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6) Powdered talc ("Mistron Vapor", Sierra Talc co., average particle size 3 μ m, maximum particle size 6 μ m)

7) Softening agent

8) Stearic acid

9) Titanium coupler

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Table 2

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Physical Properties

| | <u>E^a)₁</u> | <u>C^b)₁</u> | <u>C₂</u> | <u>E₂</u> | <u>E₃</u> | <u>E₄</u> | <u>C₃</u> | <u>C₄</u> | <u>C₅</u> |
|----------------------------|-----------------------------------|-----------------------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|
| 10 G*30 ¹⁾ | 4 | 4.2 | 5 | 3.8 | 6.7 | 7.2 | 2.1 | - | 2.9 |
| G*-10 ²⁾ | 14.7 | 16.0 | 25 | 13.6 | 23.4 | 28 | - | - | - |
| G*-10/G*30 | 3.7 | 3.8 | 5 | 3.6 | 3.5 | 3.9 | - | - | - |
| 15 G* A ³⁾ | 14.1 | 55 | 25.2 | - | - | - | - | - | - |
| Tan δ 30 ⁴⁾ | 0.54 | 0.59 | 0.69 | 0.58 | 1.3 | 1.0 | 0.3 | - | 0.34 |
| 20 Tan δ -10 ⁵⁾ | 0.83 | 0.82 | 0.98 | 0.89 | 1.3 | 1.1 | - | - | - |
| CMV ⁶⁾ | +5% | -85% | +40% | - | - | - | - | - | - |

25 a) Example

b) Comparative Example

1) G* (Kg/cm²) measured at 30 °C2) G* (Kg/cm²) measured at -10 °C3) G* (Kg/cm²) measured at -10 °C where the rubber sample had been aged for 16 hours at -10 °C

4) Tanδ measured at 30 °C

30 5) Tanδ measured at -10 °C

6) Rate of change of the Mooney viscosity before and after an aging for 168 hours at 100 °C

Claims

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1. A rubber composition having a complex shear elastic modulus of 1-20 kg/cm² and a Tan δ of 0.40-1.5 at 30° under a frequency of 0.5 Hz ± 50 % comprising:

a) x parts by weight of a filler for use in rubber;

b) y parts by weight of carbon black having an iodine adsorption of 20-120 mg/g; and

40 c) 100 parts by weight of polyisobutylene;

wherein the value x and y satisfy the relation that:

x ≤ 450, y ≤ 150, and

x + y ≥ 100.

- 45 2. A rubber composition according to claim 1 wherein said polyisobutylene has a molecular weight not less than 300,000.

3. A rubber composition according to claim 1 wherein said polyisobutylene has a molecular weight not less than 1,000,000.

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4. A rubber composition according to any one of claims 1 to 3 further comprising at least one of the following rubber components, butyl rubber, halogenated butyl, natural rubber, polyisoprene, styrene-butadiene rubber, acrylonitrile-butadiene rubber, chlorosulfonated polyethylene rubber, ethylene propylene rubber, chloroprene rubber, butadiene rubber, or polynorbornane rubber.

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5. A rubber composition according to claim 4 which is vulcanized by addition of vulcanizing agents or vulcanizing accelerators.

6. A rubber composition as claimed in any one of claims 1 to 5 having a complex shear elastic modulus of 3-12 kg/cm².
7. The use of a rubber composition as claimed in any one of claims 1 to 6 in the manufacture of a damper.
8. The use as claimed in claim 7 wherein the damper is for use in structures, public works, vehicles, oversize powered machines, sound products, meters, or vehicle components.
9. A damper comprising a rubber composition as claimed in any one of claims 1 to 6.

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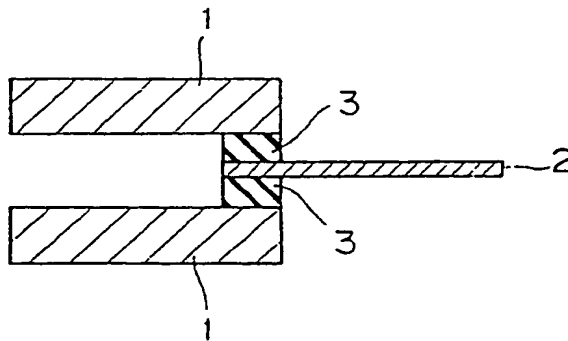
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Fig. 1



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21.11.91 Bulletin 91/47

(84) Designated Contracting States :
DE FR GB IT

(88) Date of deferred publication of search report :
07.01.93 Bulletin 93/01

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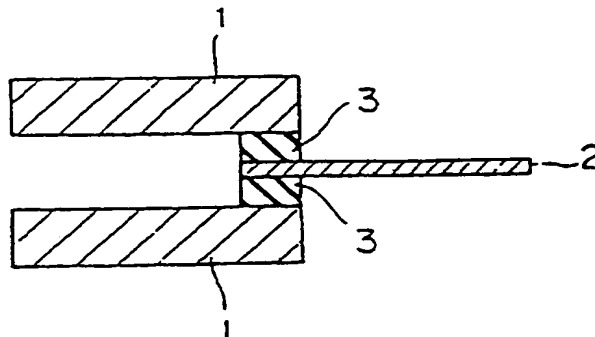
a) x parts by weight of a filler for use in rubber;

b) y parts by weight of carbon black having an iodine adsorption of 20-120 mg/g; and

c) 100 parts by weight of polyisobutylene; wherein the value x and y satisfy the relation that:

$x \leq 450$, $y \leq 150$, and
 $x + y \geq 100$.

Fig. 1





European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 30 4330

| DOCUMENTS CONSIDERED TO BE RELEVANT | | | |
|--|---|--|---|
| Category | Citation of document with indication, where appropriate, of relevant passages | Relevant to claim | CLASSIFICATION OF THE APPLICATION (Int. Cl.5) |
| X | EP-A-0 049 175 (FORD MOTOR COMPANY) Abstract * page 3, line 28 - page 4, line 23 * * page 8, line 4 - line 30 * * page 10, line 1 - line 8 * * page 11, line 29 - page 12, line 1 * --- | 1-9 | C08L23/22 F16F7/00 |
| A | DATABASE WPIL Section Ch, Week 8237, Derwent Publications Ltd., London, GB; Class A12, AN 82-78159E & JP-A-57 128 737 (BRIDGESTONE TIRE) 10 August 1982 * abstract * ----- | 1-9 | |
| | | | TECHNICAL FIELDS SEARCHED (Int. Cl.5) |
| | | | C08L F16F |
| The present search report has been drawn up for all claims | | | |
| Place of search THE HAGUE | | Date of completion of the search 11 NOVEMBER 1992 | Examiner R. E. Goovaerts |
| <p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p> | | | |

EPO FORM 1503 (01.81) (P.0401)